ACIDIC PRECIPITATION IN ONTARIO STUDY

WATER QUALITY CHANGES IN SUDBURY AREA LAKES, 1974-76 TO 1981-83

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SUMMARY

A survey was carried out during the summers of 1981-83 to re-sample 209 Sudbury area lakes originally sampled in 1974-76. Observed water quality changes included increases in pH and alkalinity and decreases in SO_4, Ni and Cu concentrations. The degree of observed changes showed a general relationship to distance from the Sudbury smelters indicating that reduced contaminant deposition from Sudbury sources was responsible for the observed improvements. Changes occurred primarily during the period between the two surveys (1976-81), not within either survey period. Although within a set of acidic lakes, small, consistent, annual increases in pH and decreases in SO_4 concentrations were found during the recent survey (1981-83), between-year differences were not statistically significant, indicating that if water quality improvements are continuing, the annual rate of change is sufficiently small to be masked by natural fluctuations.

Although changes in water quality have occurred in many Sudbury area lakes over the course of this study, and there are indications of improvements in the fisheries of some, many lakes remain acidic and metal-contaminated. During 1981-83, 16% of the study lakes continued to exhibit an average pH<5.5, 15% had average alkalinities ≤ 0, and 7% and 22% showed average concentrations of Ni and Cu, respectively, which exceeded M.O.E. objectives for the protection of aquatic life. In comparison, during 1974-76, 19% of the lakes exhibited average pH <5.5, 22% had average alkalinities ≤ 0 and 8% and 62% had average concentration of Ni and Cu respectively, exceeding M.O.E. objectives.

If ≤ 0 total inflection point alkalinity is taken as the criteria defining "acidified" lakes, no study lakes which were not acidified in 1974-76 have become acidified since that time.

INTRODUCTION

Adverse impacts on terrestrial (Gorham and Gordon 1960; Whitby et al., 1976; McIlveen and Balsillie 1978; and others) and aquatic (Gorham and Gordon 1960, 1963; O.W.R.C. 1970; Beamish and Harvey 1972; Sprules 1975; Kwiatkowski and Roff 1976; and others) systems resulting from airborne emissions from the Sudbury smelting industry have long been recognized. During the period 1974-76, a survey of 209 lakes (Figure 1) within a ~260 km radius of Sudbury, was conducted by the Ontario Ministry of the Environment (M.O.E.) to document atmospheric influences on lake waters on a regional basis. That study (Conroy et al. 1978) identified a large zone of low pH (<5.5) lakes extending northeast-southwest of Sudbury. The low pH zone, in which substantial loss of fish populations was evident, occupied an area of ~5,300 km^2 containing a lake surface area of ~650 km^2. Elevated concentrations of Ni and Cu observed in many Sudbury area lakes posed a further concern relative to potential adverse impacts on aquatic biota.

Since the mid-1970's, a number of factors including smelting process changes, pollution abatement measures and extended strike and shutdown periods in the Sudbury metal recovery industry have resulted in substantially reduced airborne emissions (O.C.T.F. 1982). During the period 1978-81, estimated annual SO_2 emissions from the Sudbury smelters ranged from 1359 to 2562 tonnes/day while during 1970-77 annual SO_2 emissions ranged from 3663 to 6383 tonnes/day. Prior to 1970, SO_2 emissions were generally even higher, ranging from 4240 to 7034 tonnes/day for the period 1960-1969 (O.C.T.F. 1982).

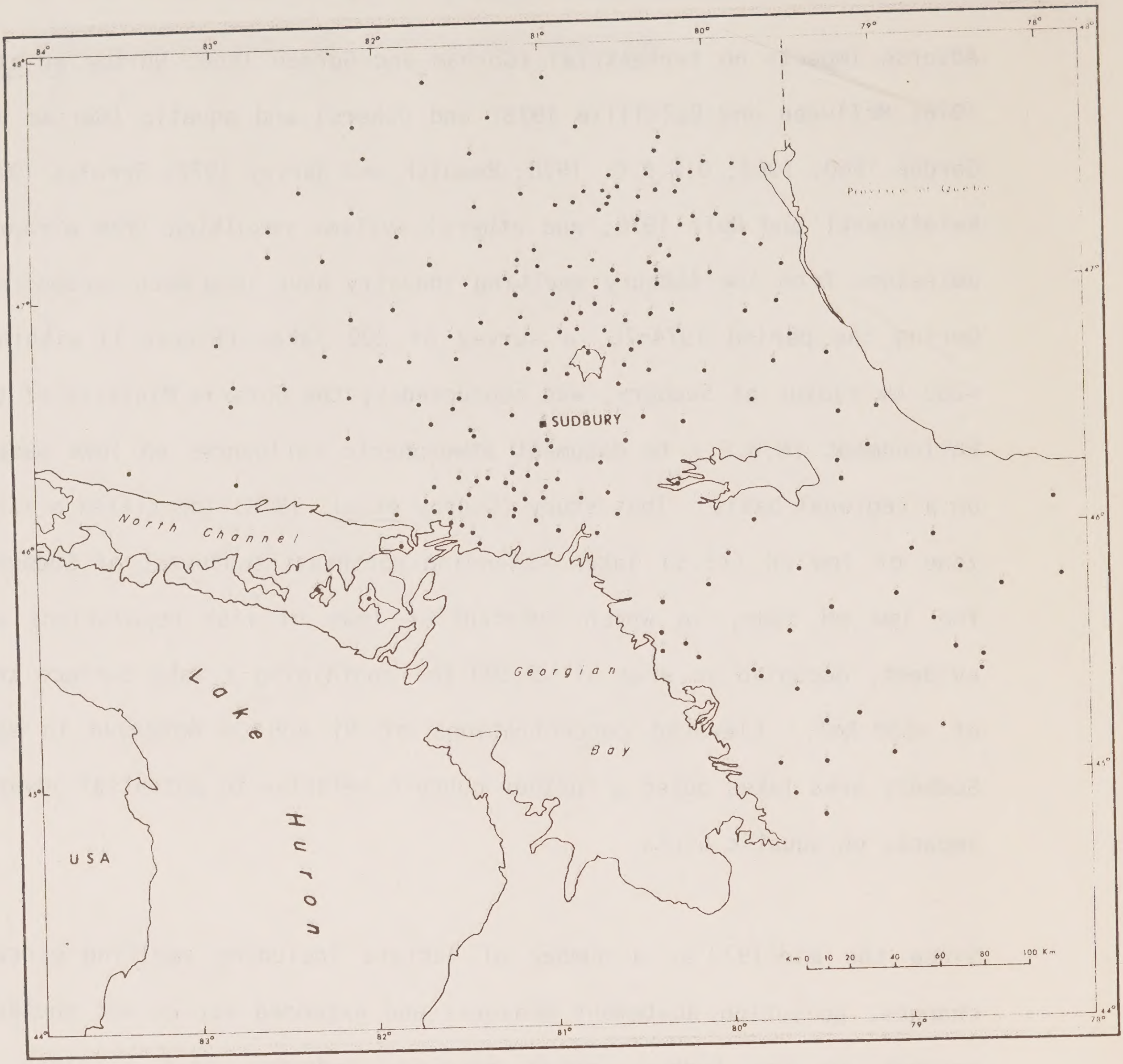

Figure 1. Locations of the study lakes.

During the summers of 1981-83, re-sampling of the lakes previously studied in 1974-76 was carried out under the Acidic Precipitation in Ontario Study (A.P.I.O.S.). This report examines changes in water quality between the study periods.

METHODS

Sampling

In order to conform with current lake sampling procedures employed under the overall A.P.I.O.S. study, the sampling format in 1981-83 differed from that used in 1974-76.

During 1981-83, each lake was sampled once at a central location between mid-June and mid-August in each of the study years. Water samples were collected as non-volume-weighted water column composites by the tygon tube method (M.O.E. 1979). They were generally taken to the lower limit of the metalimnion, defined as the depth below the region of greatest temperature change at which the observed temperature decrease was less than or equal to 0.5°C/m. Points of inflection of lake temperature profiles and respective temperatures were measured with a YSI Model 43TD telethermometer. In lakes too shallow to develop complete thermal stratification, samples were taken to 1 m above the lake bottom.

During 1974-76, samples were collected as surface grabs between May and October of each study year (Conroy *et al*. 1978). To ensure temporal comparability with the 1981-83 results, only data collected during June

to August are considered herein. The resulting data base comprised 85 lakes sampled in all three years, 82 lakes sampled during each of two years and 42 lakes sampled only during one year. Overall, 73% of the lakes were sampled 3-6 times, 24% were sampled twice, and only 3% were sampled once.

The representativeness of the data was assessed by comparison of our 1974-76 averages with average values from other, more frequent (n = 6-10) samplings available for some of the lakes for the same time period (Keller unpublished data). For a group of eight lakes, the average difference was 0.2 units, 12 µeq/L, 10 µeq/L, 4 µg/L and 2 µg/L for pH, alkalinity, SO_4, Cu and Ni, respectively. Comparative data for the 1981-83 results were only available for pH and alkalinity. Comparisons for 21 lakes were made between our data for two individual study years (n = 1 per year) and more frequent data (n = 5 per year; Keller unpublished data) for the same years, and between our three-year (1981-83) average (n = 3) and a three-year (1979-82) average based on 15 samples. In all cases, the average difference was less than 0.2 pH units and 5 µeq/L alkalinity. Given that our point in time data were restricted to summer samples while the comparative data were collected throughout the year, the good agreement indicated that our results were representative of general lake conditions.

The possible complication of different sampling methods was addressed by examining 1983 data from simultaneously collected surface and composite samples for a subset (n = 43) of the study lakes to ensure comparability. Comparison of surface and composite sample data did not

reveal any significant differences for the parameters considered herein ($p>0.1$ based on a paired _t_ test), and tube and surface values for all parameters were highly correlated ($r^2 \geq 0.97$; $p <0.01$; slope 0.93 - 0.99) permitting direct comparison of data collected by the two methods.

Containers

Samples for Ni and Cu analyses were collected in 500 ml acid-washed polyethylene bottles (and were subsequently preserved with HNO_3) in all years, and with the exception of 1974, containerization was consistent for SO_4 samples (500 ml polystyrene bottles). One litre soft glass bottles were used for SO_4 samples in 1974; however, the difference in containers did not appear to influence values since there were no significant differences ($p >0.1$ based on a paired _t_ test) between 1974 results and 1975 or 1976 data for our lake set.

Samples for pH and alkalinity determinations were collected in soft glass bottles in 1974-76. Although storage in soft glass may increase the pH and alkalinity of water samples with time (F. Tomassini, Water Quality Section, personal communication), testing showed that samples were not affected during the short storage period (<12 hours) prior to analysis (Conroy _et al_. 1978). During 1981-83, samples were collected in 500 ml polystyrene bottles, which do not affect pH or alkalinity during short storage periods such as that (~24 hours) of the 1981-83 samples (F. Tomassini, personal communication; Keller unpublished data).

Chemical Analyses

Measurement of pH and alkalinity was conducted at the Sudbury M.O.E. laboratory, except during 1974, during which pH was measured in the field. Laboratory pH measurements were adopted after testing showed that pH was stable during the storage period required for transportation to the laboratory (Conroy et al. 1978). During all study years, pH was measured by meter, after appropriate buffer calibration. Alkalinity was determined by titration with 0.010 N or 0.020 N H_2SO_4 to a fixed endpoint (pH 4.5) in 1974-76 while during 1981-83 more meaningful (Dillon et al. 1978) total inflection point alkalinity (TIA) titrations were conducted. Total fixed endpoint (TFA) alkalinities from 1974-76 were corrected to TIA alkalinities using the approximate theoretical conversion of TFA (µeq/L) - 30.5 = TIA (µeq/L) (M.O.E. 1982 Appendix). However, in practice, this correction procedure may have greatly overestimated alkalinities in 1974-76, since actual observed differences between TFA and TIA measurements may exceed 60 µeq/L (P. Dillon, Aquatic Ecosystems Section, unpublished data).

Analyses for SO_4, Ni and Cu were conducted at the M.O.E. laboratories in Toronto, in all study years. Sulphate analyses were done by the methyl thymol blue (MTB) colorimetric method in 1974-76 (M.O.E. 1975) and by ion chromatography (IC) in 1981-83 (M.O.E. 1981a). Since the MTB method may overestimate SO_4 concentrations in highly coloured waters (Kerekes and Pollock 1983; Watt, et al. 1983) giving higher values than IC, the use of the 1974-76 results was restricted to data for lakes of low colour (<20 Hazen units) only. In all years, total Ni and Cu analyses

were conducted by atomic absorption spectrophotometry on acid digested samples (M.O.E. 1975; M.O.E. 1981a).

Data Analyses

The data base from both surveys was maintained on computer files at Laurentian University, Sudbury, and statistical analyses of the data were performed using Laurentian's computer facilities. Differences within sampling periods were evaluated with one way analysis of variance (ANOVA), differences between period averages and between individual years were evaluated with paired *t* tests and correlations between variables were examined by linear regression analyses.

RESULTS AND DISCUSSION

Water Quality Differences Between 1974-76 and 1981-83

Figure 2 is a plot of average lake pH in 1974-76 against average pH recorded in 1981-83. It is apparent from Figure 2 that pH increases with time have occurred in the more acidic study lakes (pH<5.5) which tend to be located relatively close (within ~100 km) to Sudbury. In contrast, no consistent pattern of change was evident for the near-neutral study lakes, a large proportion of which are far removed from Sudbury. Lakes which previously showed obvious impacts (ie. low pH) would be most likely to show measurable positive responses to reduced acidic inputs. In addition to pH increases, general increases in alkalinity between the two study periods were also evident,

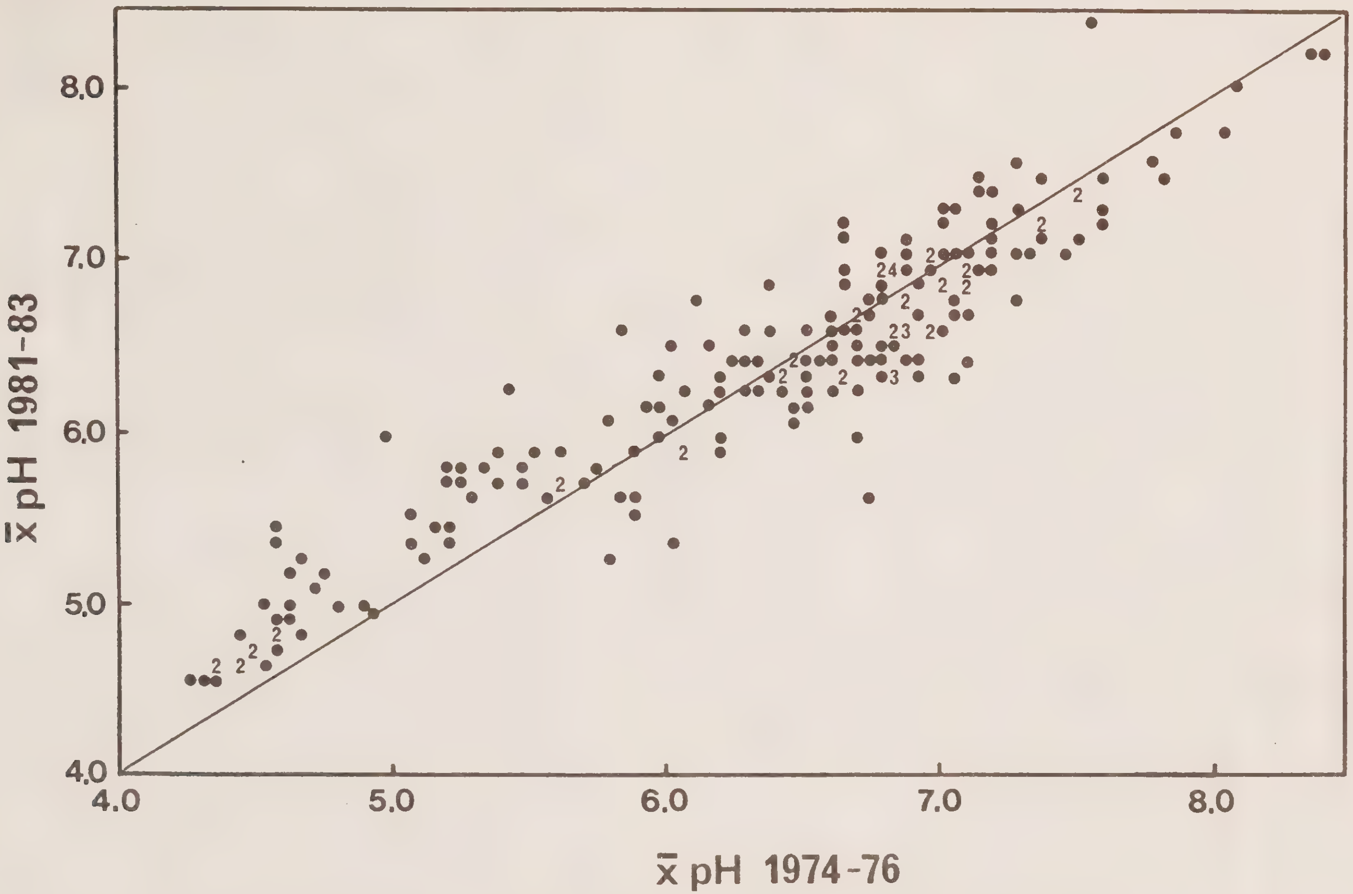

Figure 2. Average pH in 1974-76 against average pH in 1981-83 for the study lakes. The line represents a 1:1 relationship. Numbers indicate coincident points.

particularly in lakes very close (within 20 km) to Sudbury (Figure 3). It should be noted that because of our correction procedure for the 1974-76 alkalinity data (see methods), Figure 3 provides a conservative estimate of changes in alkalinity. If ≤ 0 alkalinity is taken as the definition of "acidified" (M.O.E. 1981b), 15% and 22% of the lakes would be classified as acidified during 1981-83 and 1974-76, respectively. No lake with an average alkalinity >0 in 1974-76 had an average alkalinity ≤ 0 in 1981-83.

Proximity to the emission source, reflecting the degree of previous impact, appears to exert an influence on other temporal water quality changes observed. Reductions in average lakewater concentrations of total Ni and Cu since the mid-1970's are greatest proximal to Sudbury (Figure 3) with substantial reductions over time having occurred within ~40 km for Ni and to >100 km for Cu. Nickel and Cu in the smelter emissions seem to be present predominantly in a similar size fraction (>2.5 μ; Chan *et al.* 1982a, 1982b); however, Cu apparently exhibits greater dispersion than Ni (Figure 3). Although Ni and Cu emissions are of similar magnitude (Ozvacic 1982), at distances greater than ~20 km from Sudbury, Cu deposition is comparatively more important than Ni deposition. In comparison to south-central Ontario, Ni and Cu deposition showed elevations of 4 and 20 fold respectively within 30 to 50 km of Sudbury (Jeffries 1982). Based on metal ratios in atmospheric precipitation and lake sediments in the Sudbury area, Semkin and Kramer (1976) also suggested a deposition sequence of Ni before Cu, agreeing with our observations on relative concentration patterns in lake waters.

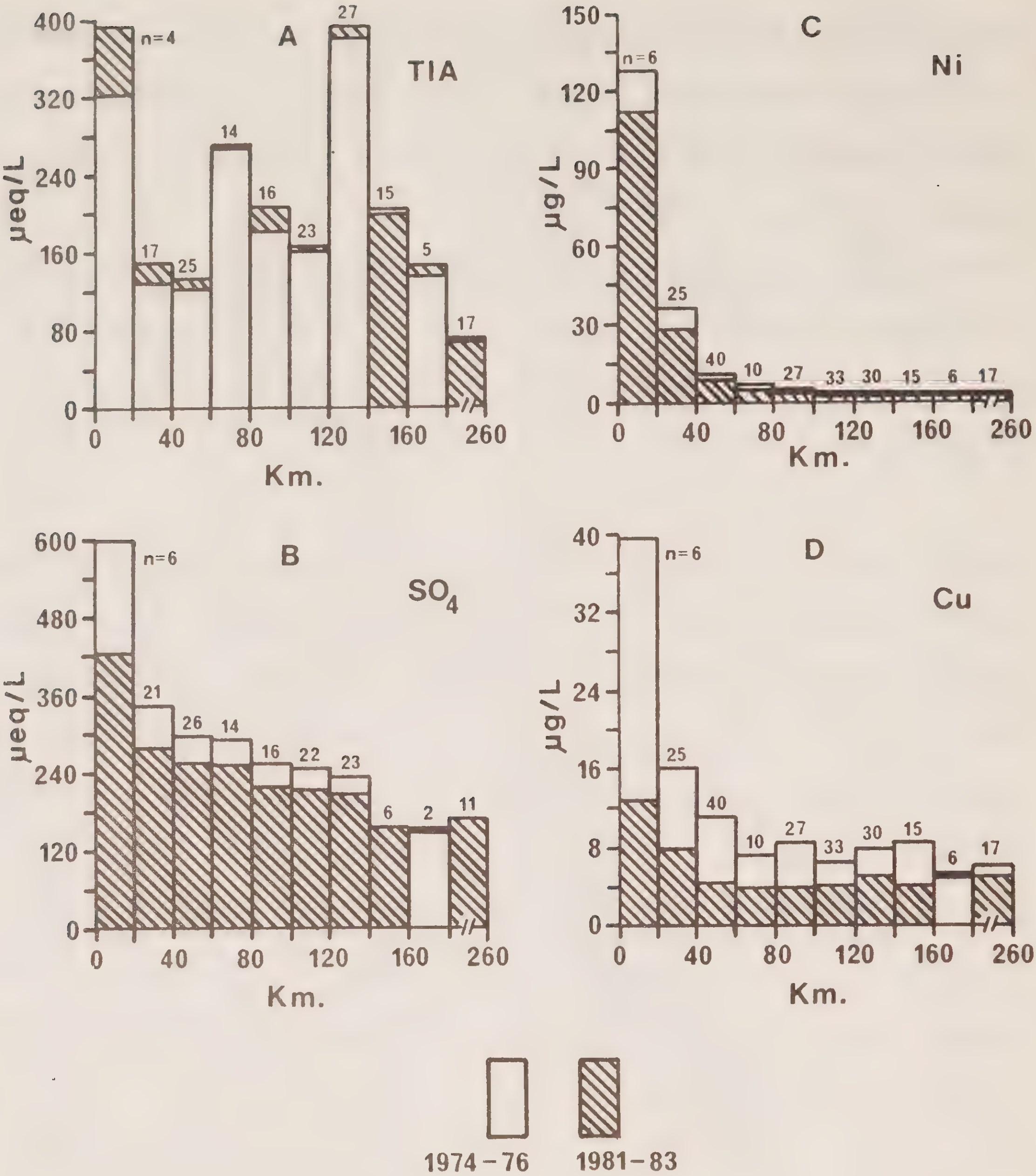

Figure 3. Average alkalinity (A) and concentrations of SO_4 (B) Ni (C) and Cu (D) by 20 km distance interval for the study lakes based on 1974-76 and 1981-83 data. Total inflection point alkalinity (TIA) for 1974-76 calculated from measured fixed endpoint alkalinity (TFA) by subtracting 30.5 μeq (M.O.E. 1982, appendix) from the 1974-76 TFA values. Number of lakes within each distance category is indicated. The figure includes only lakes with positive alkalinity for TIA, only lakes with colour <20 Hazen units for SO_4, and all lakes for Ni and Cu.

The fact that temporal changes in lakewater Ni concentrations were proportionally less than changes in Cu may reflect greater movement of Ni through watersheds to the aquatic environment. Data from 11 Sudbury area watersheds, (Jeffries et al. 1982), showed generally higher retention coefficients for Cu than for Ni. In some cases, negative retention coefficients for Ni indicated that watersheds were a Ni source. Dillon (1982), showed that during the eight-month Inco shutdown in 1978-79, Cu inputs to five Sudbury area lakes were reduced. Only one of the lakes, for which direct deposition to the lake surface was the major Ni source, showed a reduction in Ni inputs. Fitchko (1978) has indicated that in the Sudbury area, Ni is more mobile in terrestrial and aquatic environments than Cu. Our observed reductions in waterborne trace metal concentrations seem largely attributable to reduced atmospheric deposition, although in lakes showing increases in pH, reduced metal solubility at higher pH must also be involved in controlling waterborne metal concentrations. Based on relative solubilities and complexation characteristics, Stokes (1981) has suggested that Ni would be expected to persist longer than Cu in aquatic environments.

Changes in concentrations of SO_4 also showed patterns related to distance from Sudbury (Figure 3). Lakewater SO_4 concentrations were strongly distance dependent, with elevated concentrations (>200 μeq/L) occurring to >100 km from Sudbury. Substantial SO_4 reductions over time have occurred within this zone.

Consideration of relationships between observed changes in water chemistry and physical lake characteristics for a subset (n = 21) of low pH (pH<5.5 in 1974-76) study lakes, which exhibited significant changes between 1974-76 and 1981-83 (Table 1), provides further evidence that observed changes in chemistry relate to reduced impacts by Sudbury area emissions. Significant linear correlations ($r=0.5-0.7; p<0.05$) between decreases in concentration (difference between 1974-76 and 1981-83 averages) and proximity to the smelters were found for SO_4, Ni, and Cu. A lack of significant correlations ($p>0.1$) between watershed area/lake area ratio and changes in pH, Ni and Cu indicated that direct deposition to lake surfaces may be very important for H^+ and trace metals. A weak correlation ($r=0.3; p<0.1$) between decreases in SO_4 concentrations and increasing watershed area/lake area ratio suggested that both atmospheric and terrestrial pathways may be important for SO_4.

Additional evidence exists which indicates that recent improvements in water quality have occurred in Sudbury area lakes. Clearwater Lake, a very intensively studied lake close (13 km) to the Sudbury smelters (Dillon *et al*. 1979) has shown increased pH and declines in SO_4 and trace metal concentrations since the mid-1970's (Figure 4).

Chronology of Observed Changes

Although improvements in water quality, related to reduced smelter emissions, are evident in many Sudbury area lakes, determination of exact cause-effect relationships is not possible, since numerous

TABLE 1. Annual and sampling period averages of pH, SO_4, Cu and Ni for 21 study lakes which exhibited average pH <5.5 in 1974-76.

	pH	SO_4 (μeq/L)	Cu (μg/L)	Ni (μg/L)
Annual Averages				
1974	4.51	336	24	38
1975	4.66	321	15	44
1976	4.72	339	13	51
1981	4.86	294	7	36
1982	4.89	282	9	37
1983	4.92	277	7	37
Period Averages				
$\bar{x}$ 1974-76	4.62	332	17	44
$\bar{x}$ 1981-83	4.89	284	8	37
$\bar{x}$ Δ	0.3 unit	-14%	-53%	-16%
Significance [a]	$p<0.05$	$p<0.05$	$p<0.01$	$p>0.1$

[a] significance of differences between period averages based on a paired *t* test

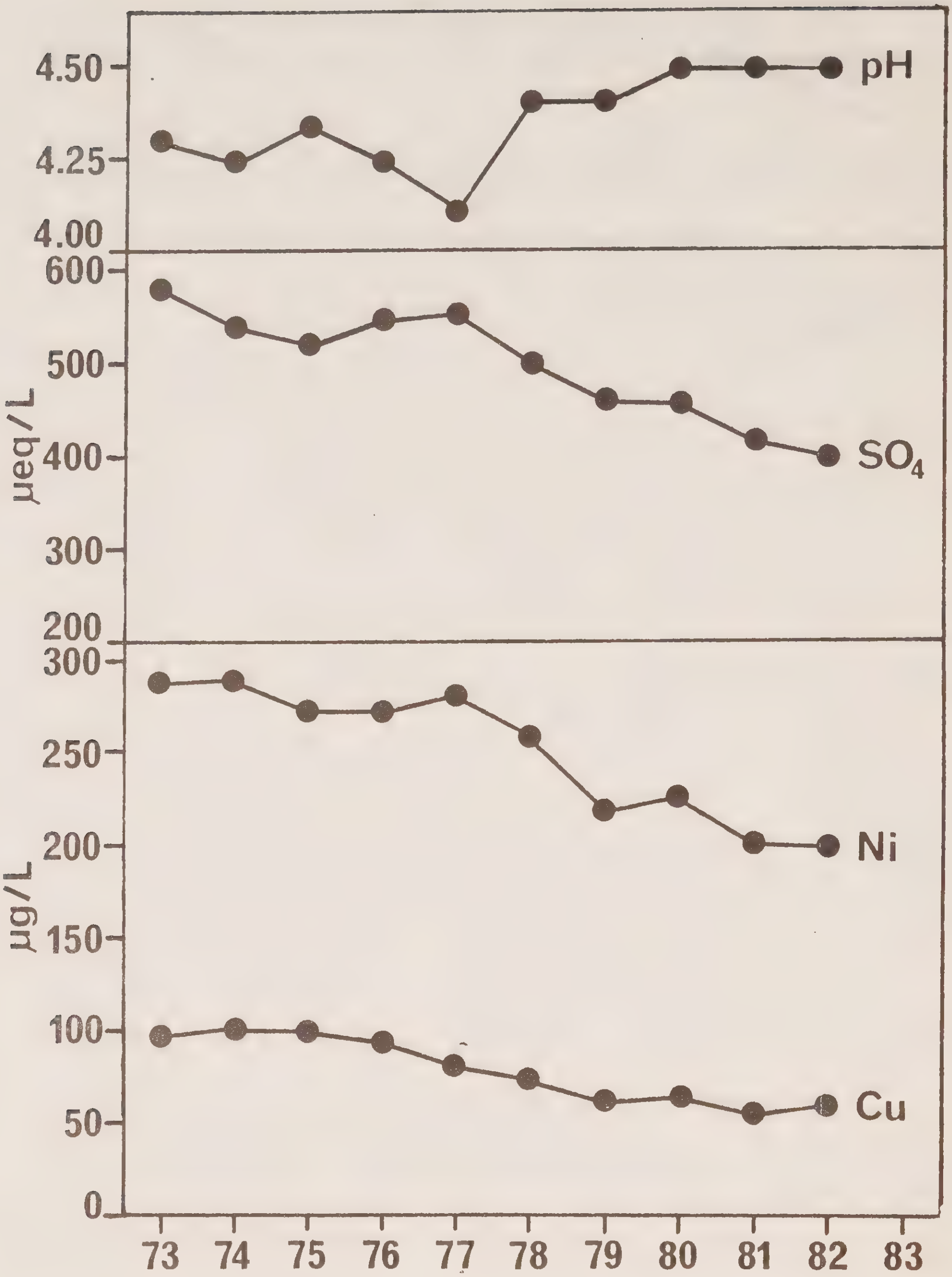

Figure 4. Temporal changes in chemistry in Clearwater Lake. Data for 1973-79 from Yan and Miller (1982); 1980-82 from P. Dillon, unpublished data.

factors including strikes, vacation shutdowns and abatement measures have contributed to generally reduced contaminant emissions since 1978. Additionally, given the often long lag time in lake responses due to the influence of hydrology and watershed effects, some recent improvements in water quality may at least partially stem from emission reductions in the early 1970's.

Averaged data for the subset of acidic lakes (Table 1) indicated that substantial changes occurred between 1974-76 and 1981-83, with significant ($p<0.05$) differences in average pH, Cu and SO_4 between the periods. Although, on average, Ni concentrations showed temporal reductions within 40 km of Sudbury (Figure 3), differences were not statistically significant ($p>0.1$). Examination of the data showed that with the exception of a significant ($p<0.05$) decrease in Cu between 1974 and 1975, which may be related to the commissioning of the 381 m Inco "Superstack" in 1972 and general reductions in emmissions at the beginning of the decade, no statistically significant ($p<0.05$) differences in concentrations occurred between individual years within either sampling period. This suggested that small annual changes may be masked by natural fluctuations. Nevertheless, it was noted that, on average, small annual increases in pH and decreases in SO_4 occurred throughout the recent study (1981-83; Table 1).

Inspection of Table 1 indicates that changes occurred primarily during the period between the two surveys (ie. 1976-1981). In addition to short-term vacation and production shutdowns, major events which probably contributed substantially to reduced Sudbury area emissions

during that period include 1) the eight-month Inco strike and shutdown in 1978-79, 2) implementation of a new smelting process, with increased sulphur containment, at Falconbridge in 1978, 3) closure of the pelletizing plant associated with the Inco Iron Ore Recovery Plant in 1980, 4) installation of additional electrostatic precipitators on the Iron Ore Recovery Plant 194m stack in 1980 to reduce particulate emissions, and 5) implementation of a 1980 regulation restricting the Inco smelter to average SO_2 emissions of 2500 tonnes/day.

Implications for Aquatic Biota

The reduced concentrations of Ni and Cu in many of the study lakes may be of considerable biological importance since alone, or in combination, high concentrations of these metals may be toxic to aquatic biota, particularly in association with low pH. Comparison of average lake trace metal concentrations during the two surveys with M.O.E. objectives (M.O.E. 1978) for protection of fish and aquatic life in surface waters indicated that only in a relatively small proportion of lakes did average Ni and Cu (7% and 22% respectively) concentrations exceed objectives (25 and 5 µg/L for Ni and Cu, respectively) during 1981-83, while a larger proportion of average trace metal concentrations were in exceedence in 1974-76 (8% and 62% for Ni and Cu respectively).

The extent to which trace metal toxicity was implicated in observed fish population losses during the acidification of Sudbury area lakes is not known. However, assuming that metal emissions are roughly proportional to SO_2 emissions, it is likely that trace metal toxicity played a large role in the observed fishery impacts, since during the period when most

fishery losses occurred, 1950-70 (Beamish and Harvey 1972; Gunn 1982), trace metal emissions were probably 2 to 4 times present levels (based on consideration of historical SO_2 emmissions data presented in O.C.T.F. 1982). Additionally, lake pH's at the time of fishery losses may have been substantially less than pH's recorded in the last decade. Reliable measurements from the 1950's and early 1960's do not exist; however, comparison of pH data (field-meter measurements) from 1969-70 (O.W.R.C. 1970) with our 1974-76 data for 19 low pH (<5.5) Sudbury area lakes showed a significant ($p<0.05$) increase in pH, on average, for the more recent measurements ($\bar{x}$ 4.5 in 1969-70; $\bar{x}$ 4.8 in 1974-76).

Information on fish communities in affected Sudbury area lakes is unfortunately scarce; however, there are indications of improvement in the fish populations of some lakes in response to pH increases/metal concentration decreases.

Joe Lake, 28 km N of Sudbury, currently supports a self-sustaining brook trout (*Salvelinus fontinalis*) population resulting from introductions in 1977 (J. M. Gunn unpublished data). The natural brook trout population of Joe Lake disappeared during the late 1950's or early 1960's and stockings of the species in 1959 and 1966 were unsuccessful (Ontario Ministry of Natural Resources (M.N.R.) unpublished data). Available chemistry data for Joe Lake were not collected by identical methods, but nevertheless a substantial increase in pH and decrease in trace metal concentrations with time is evident (Table 2).

TABLE 2. Temporal comparison of available pH and trace metal data for Joe Lake.

	pH	Cu (μg/L)	Ni (μg/L)
1973[a,b]	4.8[a], 5.0[b]	42[b]	-
1975[c]	5.7	20	23
1976[c]	5.6	17	25
1981[d]	6.3	3	14
1982[d]	6.3	10	14
1983[d]	6.2	2	10

[a] J. R. Kramer unpublished data; surface sample in summer.

[b] Chau and Lum-Shue-Chan (1974); surface sample in summer; total metal concentration given is the sum of "labile" and "strongly bound" values reported.

[c] M.O.E. 1982 (appendix); ice free period average for whole lake volume weighed samples.

[d] present study.

Whitepine Lake, 90 km N of Sudbury until very recently contained only a remnant native lake trout (*Salvelinus namaycush*) population consisting of a very small number (~15) of large adults. During 1982 (lake pH ~5.7), native lake trout fingerlings were observed, the first evidence of recruitment documented in the lake during the last decade (Gunn and Keller 1984). Historical chemistry data are not available for Whitepine Lake. Additional anecdotal information (M.N.R. unpublished data; conversations with local residents) suggesting improvement in fisheries exists for other Sudbury area lakes including Bigwood (80 km N), Bluesucker (83 km NE), Kukagami (48 km NE) and Seagram (78 km NE). The current pH of these lakes ranges from 5.2-5.8 (present study).

Although such examples indicate that positive biological responses to improved water quality have occurred in some Sudbury area lakes, a large number of lakes continue to exhibit water quality which probably exerts a strong negative impact on aquatic biota, fish in particular. Fifteen percent of the study lakes may still be defined as acidified (alkalinity ≤ 0), and as indicated previously, concentrations of Ni and Cu exceeding M.O.E. objectives for the protection of aquatic life were recorded in many of the lakes.

CONCLUSIONS

Many lakes in the greater Sudbury area have shown improvements in water quality between 1974-76 and 1981-83. Observed increases in pH and alkalinity and decreases in SO_4 and trace metal concentrations reflect reduced contaminant deposition from the Sudbury smelting industry, since patterns of change bear general relationships to distance from the Sudbury smelters. The greatest changes in chemistry occurred between 1976 and 1981, representing the combined influence of improved abatement measures, stricter government requirements and emission reductions due to extended non-operating periods.

Some Sudbury area lakes show signs of recent improvement in their fish populations, apparently resulting from the observed water quality improvements; however, 15% of the lakes studied remain acidic (alkalinity ≤ 0) and 7% and 22% have Ni and Cu concentrations, respectively, exceeding objectives (M.O.E. 1978) for the protection of aquatic life.

Due to the constantly changing nature of the Sudbury smelting industry during the last decade, specific cause-effect relationships between abatement efforts and lake responses cannot be established with the present data base. The results do however, emphasize the resiliency of aquatic systems, and indicate that reductions in contaminant emissions are manifested as improvements in water quality.

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